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# Search for Nova Presolar Grains: $\gamma$-Ray Spectroscopy of $^{34}$Ar and its Relevance for the Astrophysical $^{33}$Cl($p$,$\gamma$) Reaction

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The discovery of presolar grains in primitive meteorites has initiated a new era of research in the study of stellar nucleosynthesis. However, the accurate classification of presolar grains as being of specific stellar origins is particularly challenging. Recently, it has been suggested that sulfur isotopic abundances may hold the key to definitively identifying presolar grains with being of nova origins and, in this regard, the astrophysical $^{33}\mathrm{Cl}(p,\gamma)^{34}\mathrm{Ar}$ reaction is expected to play a decisive role. As such, we have performed a detailed $\gamma$-ray spectroscopy study of $^{34}$Ar. Excitation energies have been measured with high precision and spin-parity assignments for resonant states, located above the proton threshold in $^{34}$Ar, have been made for the first time. Uncertainties in the $^{33}\mathrm{Cl}(p,\gamma)$ reaction have been dramatically reduced and the results indicate that a newly identified $\ell = 0$ resonance at $E_r = 396.9(13)$ keV dominates the entire rate for $T = 0.25$–0.40 GK. Furthermore, nova hydrodynamic simulations based on the present work indicate an ejected $^{32}\mathrm{S}/^{33}\mathrm{S}$ abundance ratio distinctive from type-II supernovae and potentially compatible with recent measurements of a presolar grain.

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Cataclysmic variables (CVs) are interacting binary systems that comprise a main sequence star and a white dwarf star in close proximity. In these scenarios, the gravitational pull of the white dwarf distorts the main sequence star, allowing matter to be transferred to the compact object from its companion. This leads to irregular increases in the brilliance of the system and the brightest manifestations occur when a thermonuclear runaway is achieved on the surface of the white dwarf star [1]. Such events, known as classical novae, attain peak luminosities of $\sim 10^4$–$10^5$ $L_\odot$ and expel $\sim 10^{-7}$–$10^{-4}$ $M_\odot$ of material into the interstellar medium at high velocities ($\geq 1000$ km s$^{-1}$) every $10^4$–$10^5$ yrs, making them some of the most frequent and violent stellar explosions to occur in our Galaxy.

Over the past three decades, optical, ultraviolet (UV) and infrared (IR) observations have produced a wealth of information on the chemical composition of nova ejecta. In particular, they have revealed that the global content of metals is larger than solar [2], and the exact nature of the explosion depends strongly on the underlying composition of the white dwarf star. For example, in more massive Oxygen-Neon (ONe) novae, which achieve peak temperatures $\sim 0.4$ GK, it is expected that elements up into the Si-Ca mass region are formed [3]. However, despite the profusion of observational data available, a number of key stages of nova nucleosynthesis are still not fully understood, owing to large uncertainties in several experimentally unconstrained nuclear physics processes that govern the pathway of nucleosynthesis in such environments. Consequently, significant discrepancies still exist between theoretical models and the latest astronomical observations.

Recently, the isolation of presolar grains in primitive meteorites has opened a new window of discovery for observational astronomy [4,5]. These microscopic grains of meteoritic stardust predate the formation of the Solar System and exhibit highly anomalous isotopic abundances that reflect the nucleosynthetic processes of the parent star around which they were formed. Unfortunately, determining the stellar paternity of presolar grains is particularly challenging. Specifically, the isotopic excesses expected from different astrophysical environments can often be ambiguous. Therefore, it is imperative that distinctive isotopic signatures, demonstrative of specific stellar origins, be identified. In this regard, it has been suggested that sulfur isotopic ratios [6–8] may hold the key to accurately identifying ONe nova presolar grains, and several experimental studies have been performed to reduce uncertainties in the proton radiative capture rates of stable sulfur isotopes [9–11]. That being said, the astrophysical reactions involving unstable nuclei, which dramatically affect the ejected abundances of sulfur isotopes in classical nova explosions, remain largely unknown. In particular, nova sensitivity studies indicate that the astrophysical $^{33}\mathrm{Cl}(p,\gamma)^{34}\mathrm{Ar}$ reaction is likely to be of special significance [12].

At present, almost no experimental information exists on the $^{33}\mathrm{Cl}(p,\gamma)^{34}\mathrm{Ar}$ reaction and previous estimates of its rate have been based solely on Hauser-Feshbach (HF) calculations [13]. These calculations are likely to be grossly inappropriate for relatively low-temperature astrophysical environments, in which only a few resonant states, located above the proton-emission threshold energy of 4663.9(4) keV in $^{34}$Ar [14], are expected to govern the entire stellar reaction rate. Consequently, the $^{33}\mathrm{Cl}(p,\gamma)^{34}\mathrm{Ar}$ reaction is currently uncertain by orders of magnitude over the temperature range of classical novae, leading to variations in the predicted ejected abundances of $^{33}$S and $^{34}$S by factors of $\sim$18 and $\sim$3, respectively [12]. Recently, a high-resolution $^{36}\mathrm{Ar}(p,t)$ reaction study to identify excited states in $^{34}$Ar was performed by Long *et al.* [15]. In that study [15], 16 excited levels were observed, but no spin-parity information was obtained. Moreover, the $^{36}\mathrm{Ar}(p,t)$ reaction selectively populates natural-parity states and, as such, it is likely that there are a number of excited states in $^{34}$Ar corresponding to key resonances in the $^{33}\mathrm{Cl}(p,\gamma)$ reaction that have yet to be identified [15]. In this Letter, we describe a measurement that employs a heavy-ion fusion-evaporation reaction to simultaneously populate excited states in the astrophysically important nucleus $^{34}$Ar and its well-studied mirror analog, $^{34}$S [16]. Mirror nuclei are expected to exhibit nearly identical structures [17] and, as such, by studying the $\gamma$ decays of both $^{34}$Ar and $^{34}$S with the advanced $\gamma$-ray tracking array GRETINA [18], we have been able to make the first firm spin-parity assignments for $^{33}\mathrm{Cl} + p$ states up to resonance energies $E_r$ of 400 keV. In particular, we have identified two $s$-wave resonances and a $p$-wave one in the Gamow-energy window of hydrogen burning for classical novae ($T_{\mathrm{peak}} = 0.2$–$0.4$ GK), herewith reducing uncertainties in the astrophysical $^{33}\mathrm{Cl}(p,\gamma)^{34}\mathrm{Ar}$ reaction by orders of magnitude. This technique has previously been successful for the study of particle-unbound levels in key astrophysical nuclei [19–22]. However, in this case, the coupling of GRETINA with the Argonne fragment mass analyzer (FMA) [23] offered a number of unique experimental advantages. Namely, it provided a much larger solid angle for recoil detection than past setups, a high efficiency for high-energy $\gamma$ rays and significant segmentation, resulting in improved Doppler reconstruction.

Here, a 15-pnA, 95-MeV beam of $^{24}$Mg ions, produced by the ATLAS accelerator, was used to bombard a $\sim$200 $\mu\mathrm{g/cm^2}$-thick $^{12}$C target for 140 h in order to produce $^{34}$Ar nuclei via the $2n$ evaporation channel. Prompt $\gamma$ rays were detected using the GRETINA tracking array which, in this instance, consisted of 12 modules, in coincidence with $A = 34$, charge state $13^+$ recoils, transmitted to the focal plane of the FMA. The focal plane position was determined with a position-sensitive parallel-grid avalanche counter (PGAC) and $^{34}$Ar, $^{34}$Cl, and $^{34}$S nuclei were cleanly resolved from $\Delta E - E$ information in a subsequent ionisation chamber, as illustrated in Fig. 1. Corresponding energy and efficiency calibrations were carried out using standard $^{152}$Eu and $^{56}$Co $\gamma$-ray sources (identical tracking conditions [24] were employed for both calibration and experimental data). A $\gamma$-$\gamma$ coincidence matrix was produced and analyzed, together with $\gamma$-ray singles data, in order to obtain information on the $^{34}$Ar and $^{34}$S decay schemes.

Table I presents the level energies, transition energies, $\gamma$-ray intensities, and spin-parity assignments for observed excited states in $^{34}$Ar, together with a comparison to previous work [15,16]. In general, good agreement is found between the presently determined $^{34}$Ar excitation energies, and Refs. [15,16]. Moreover, previously observed $\gamma$ decays at 2091.3(5), 1197.5(4), 3289.1(10), 1785.6(7), 1228.4(5), and 2424.7(22) keV from the well-known $2_1^+$, $2_2^+$, $0_2^+$ and $3_1^-$ levels in $^{34}$Ar [16] are also observed here. The tracked $\gamma$-ray singles spectrum detected in coincidence with $^{34}$Ar recoils is shown in Fig. 2, while Fig. 3 illustrates the presently deduced level schemes for $^{34}$Ar and $^{34}$S.

Examining Fig. 2, we find an intense $\gamma$-ray transition at 2552.4(8) keV. A detailed $\gamma$-$\gamma$ matrix analysis has revealed

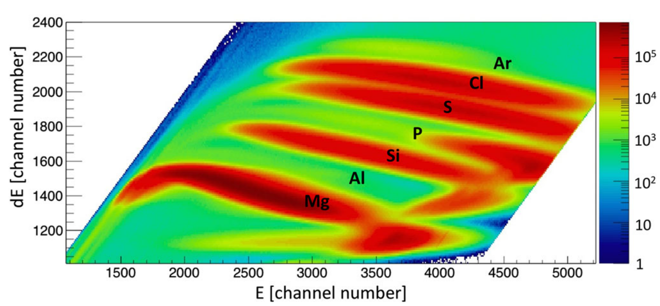


FIG. 1. An example $\Delta E - E$ plot used for $Z$ selection in the ionization chamber data. Regions associated with different atomic numbers are labeled.

TABLE I. Properties of excited states in $^{34}$Ar. Previous excitation energies have been taken from Ref. [16], unless noted otherwise. Level energies have been corrected for the recoil of the compound nucleus [22].

| $E_x$ (keV) previous [16] | $E_x$ (keV) present | $E_\gamma$ (keV) | $I_\gamma$ | $J^\pi$ |
|---|---|---|---|---|
| 2091.1(3) | 2091.4(5) | 2091.3(5) | 100(5) | $2_1^+$ |
| 3287.7(5) | 3289.0(7) | 1197.5(4) | 37.9(16) | $2_2^+$ |
| | | 3289.1(10) | 5.3(6) | |
| 3873.0(30) | 3876.2(9) | 1784.8(8) | 5.9(4) | $0_2^+$ |
| 4019.1(43) [a] | 4020.8(18) | 1930.4(23) | 4.8(9) | $2_3^+$ |
| | | 4019.8(15) | 7.4(9) | |
| 4127.8(10) | 4131.7(10) | 842.5(7) | 2.4(2) | $1_1^+$ |
| 4513.2(8) | 4517.3(10) | 1228.4(5) | 15.8(7) | $3_1^-$ |
| | | 2424.7(22) | 2.3(5) | |
| 4641.3(21) [a] | 4643.9(9) | 2552.4(8) | 25.1(16) | $4_1^+$ |
| 4865.0(40) | 4851.6(13) | 1562.8(7) | 5.8(4) | $3_1^+$ |
| | | 2759.9(12) | 11.8(9) | |
| 4875.9(38) [a] | 4881.3(21) | 1592.5(17) | 1.4(2) | $2_4^+$ |
| | | 2788.9(19) | 4.4(5) | |
| | | 4881.9(24) | 3.6(6) | |
| 4967.2(27) [a] | 4963.8(13) | 832.1(9) | 1.6(2) | $0_3^+$ |
| 4967.0(40) | 4966.7(11) | 2875.2(10) | 11.6(9) | $2_1^-$ |
| *new state* | 5060.8(13) | 1771.8(11) | 5.0(4) | $1_2^+$ |
| | | (5062) | < 1.4 | |

[a]Energy taken from $(p, t)$ reaction study of Long *et al.*[15].

that this $\gamma$ ray is coincident with the $2_1^+ \to 0_1^+$ transition, indicating an excited state in $^{34}$Ar just below the proton threshold at 4643.9(9) keV. No further $\gamma$ deexcitations were observed from this state and, in comparison with the mirror nucleus $^{34}$S in the energy range $4650 \pm 500$ keV [16], only the $4_1^+$ 4689-keV state is known to exhibit a 100% $\gamma$-decay branch to the $2_1^+$ level. Consequently, we assign a $4_1^+$, 4644-keV excited state in $^{34}$Ar.

The only remaining bound states in $^{34}$Ar to be observed in the current work appear at 4020.8(18) and 4131.7 (10) keV, respectively. Here, the 4021-keV excited state is found to exhibit a high-energy, direct-to-ground state transition, together with a further decay to the $2_1^+$ level, whereas the state at 4132 keV decays via a single, 842.5(7)-keV branch to the $2_2^+$, 3289-keV state. An inspection of the mirror nucleus $^{34}$S, in the energy region $4000 \pm 500$ keV, indicates that only the $1_1^+$ and $2_3^+$ excited levels may be considered as possible analogs. Furthermore, only the $1_1^+$ state in $^{34}$S is known to decay to the $2_2^+$ level [16]. Thus, we assign 4021- and 4132-keV excited states in $^{34}$Ar as $2_3^+$ and $1_1^+$, respectively. We also note that in the $(p, t)$ reaction study of Long *et al.* [15], the 4021-keV level in $^{34}$Ar was strongly populated, while the 4132-keV state was not observed. This would indicate further support for the present assignments.

Focusing on proton-unbound levels in $^{34}$Ar, we observe strong $\gamma$-decay transitions at 2759.9(12), 2788.9(19), and 2875.2(10) keV, as illustrated in Fig. 2. These transitions are found to exhibit coincidence relationships with the 2091-keV $\gamma$ ray in $^{34}$Ar, indicating excited states at 4851.6(13), 4881.3(21), and 4966.7(11) keV that correspond to resonances in the $^{33}$Cl + $p$ system at 187.7(14), 217.4(21), and 302.8(12) keV, respectively. Additional 1592.5(17)- and 4881.9(24)-keV $\gamma$-decay branches to the $2_2^+$ and ground state levels in $^{34}$Ar are observed from the 4881-keV excited state, and an additional 1562.8(7)-keV

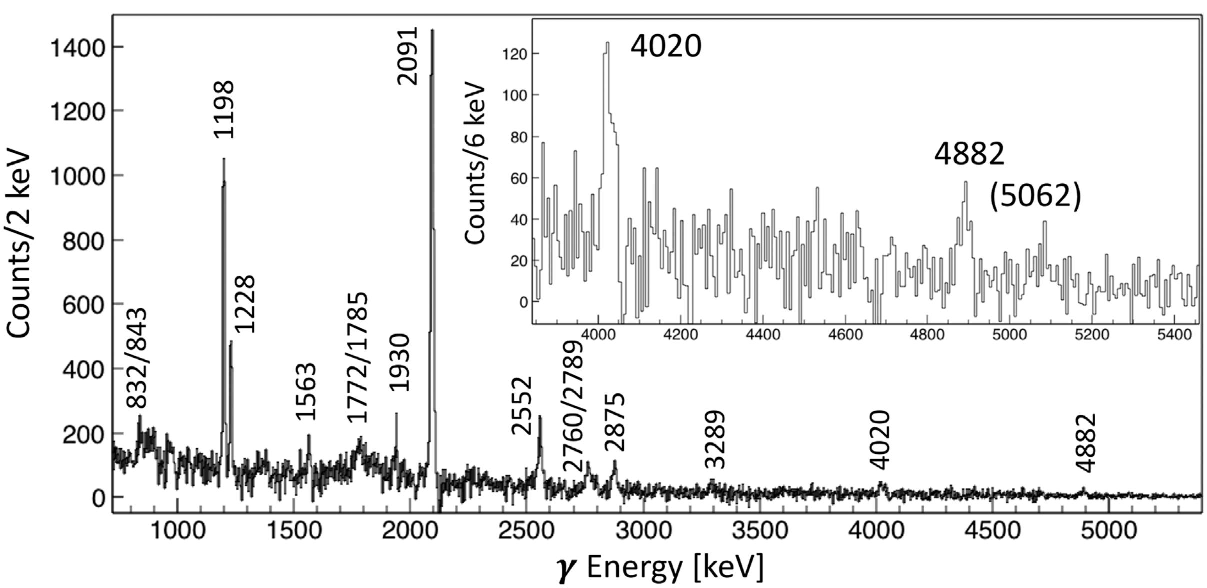


FIG. 2. Tracked $\gamma$-ray singles spectrum of $^{34}$Ar. Inset: Energy region of interest for direct-to-ground state decays of the key 4881- and 5061-keV, $\ell = 0$ resonant states.

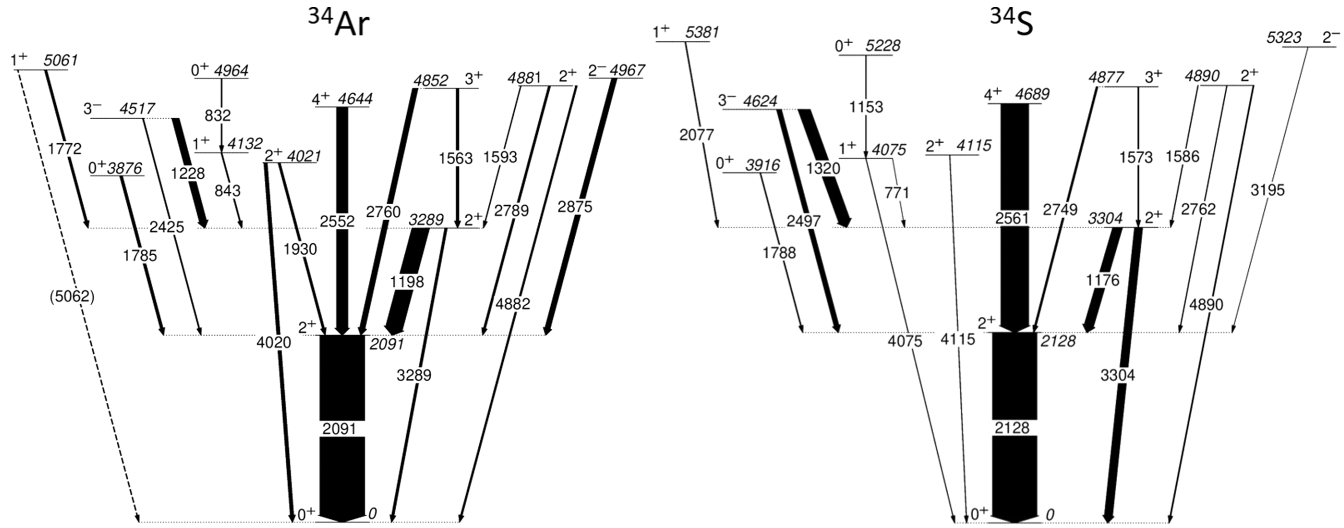


FIG. 3. Proposed level schemes of $^{34}$Ar (left) and $^{34}$S (right) based on $\gamma$ decays observed in the present study. Energies are given in keV and the widths of arrows are proportional to the relative intensities of the transitions. The level energies and $\gamma$-decay branches observed for $^{34}$S are in good agreement with previous work [16].

transition to the $2_2^+$ state is observed from the 4852-keV level. In contrast, the 4967-keV excited state was observed to only decay to the $2_1^+$ level. By examining the mirror nucleus over the energy range $4850 \pm 500$ keV, only the $3_1^+$, 4877-keV and $2_4^+$, 4890-keV excited states exhibit $\gamma$ decays to both the $2_1^+$ and $2_2^+$ levels, while the $2_1^-$, 5323-keV state is known to decay to the $2_1^+$ level with a $\sim$100% branch. The observation of a direct-to-ground state transition for the presently reported 4881-keV state rules out a $3^+$ assignment, as an M3 transition would not be observed and, thus, we assign $3_1^+$, 4852-keV, $2_2^+$, 4881-keV and $2_1^-$, 4967-keV excited states in $^{34}$Ar.

A further coincidence relationship with the $2_2^+$ excited state in $^{34}$Ar was observed in the present data at 1771.8(11) keV, as illustrated in Fig. 4, indicating a newly observed excited state at 5060.8(13) keV, corresponding to a $^{33}$Cl + $p$ resonance at 396.9(13) keV. This level was also

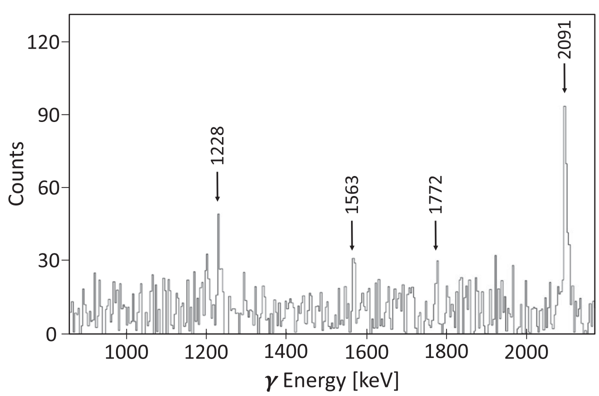


FIG. 4. $\gamma$-$\gamma$ coincidence spectrum with a gate placed on the 1198-keV, $2_2^+ \rightarrow 2_1^+$ transition.

tentatively observed to decay directly to the ground state via a high-energy $\gamma$ ray at $\sim$5062 keV, as shown in Fig. 2. Based on the mirror nucleus, only the $1_2^+$, 5381-keV and $3_2^-$, 5680-keV excited states in $^{34}$S [16] represent possible analogs. The tentative observation of a direct ground-state transition is not compatible with a $3^-$ assignment, and the relative mirror energy shift of $\sim$320 keV, as opposed to $\sim$620 keV, would strongly favor a $1^+$ assignment. Therefore, we assign the 5061-keV excited state as the $1_2^+$ level in $^{34}$Ar and, as a result, indicate a new $\ell = 0$ resonance in the $^{33}$Cl$(p,\gamma)$ reaction at 397 keV.

Finally, a low-intensity $\gamma$ ray is observed in the current study at 832.1(9) keV. This transition is likely to correspond to a $\gamma$ ray that was previously assigned as a decay to the $1_1^+$ level in $^{34}$Ar [16], indicating an excited state at 4963.8(13) keV that corresponds to a resonance in the $^{33}$Cl + $p$ system at 299.9(14) keV. The energy of this state is in good agreement with the established $0_3^+$ level in $^{34}$Ar, populated in $(p, t)$ reaction studies [15], and we adopt the same spin-parity assignment here. It should also be noted that the presently observed $\gamma$-decay path is entirely consistent with that seen in the $0_3^+$ mirror analog of $^{34}$S.

For an evaluation of the $^{33}$Cl$(p,\gamma)$ stellar reaction rate, we use our current, precise resonance energies and spin-parity assignments for states located above the proton-emission threshold in $^{34}$Ar. In particular, excited states at 4881, 5062, 4852, 4964, and 4967 keV, corresponding to $\ell = 0$ resonances at 217 and 397 keV, $\ell = 2$ resonances at 188 and 300 keV, and a $\ell = 1$ resonance at 303 keV in the $^{33}$Cl$(p,\gamma)$ reaction were considered. Spectroscopic factors, $C^2S$, for the estimation of proton partial widths, have been adopted from mirror states in $^{34}$S [25], with the exception of the 300 keV resonance (a value of 0.003 is adopted from

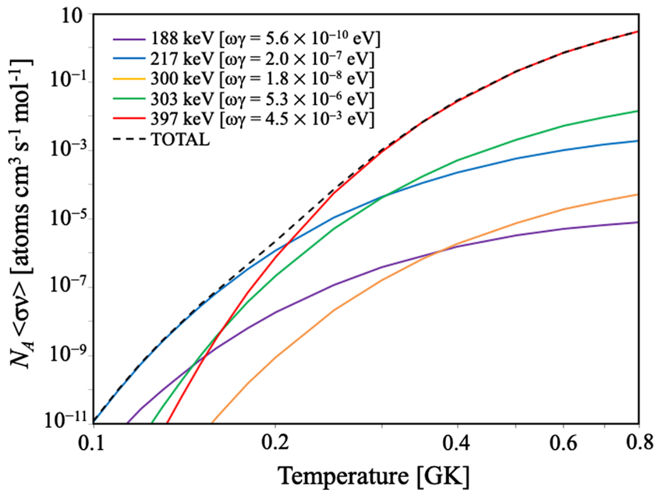


FIG. 5. Contribution of individual resonances to the $^{33}$Cl$(p,\gamma)^{34}$Ar stellar reaction rate.

shell model calculations), while $\gamma$-decay partial widths have been estimated from analog lifetimes [16] and shell model calculations. Specifically, resonance strengths of $5.6 \times 10^{-10}$, $2.0 \times 10^{-7}$, $1.8 \times 10^{-8}$, $5.3 \times 10^{-6}$, and $4.5 \times 10^{-3}$ eV have been calculated for the states at 188, 217, 300, 303, and 397 keV, respectively (we estimate an overall factor ~2 uncertainty for calculated resonance strengths).

Figure 5 shows the contribution of individual resonances to the astrophysical $^{33}$Cl$(p,\gamma)$ reaction. As can be seen, the newly identified $1^+$, $\ell = 0$ resonance at 397 keV governs the rate over almost the entire temperature range of classical novae, with the $2^+$, 217-keV, $\ell = 0$ resonance making up the dominant contribution at lower temperatures (~0.2 GK). We note here that, although a $1^+$ assignment has been adopted for the 397-keV state in this study, the alternative $3^-$ assignment would lead to a nearly identical contribution to the $^{33}$Cl$(p,\gamma)$ stellar reaction rate. Consequently, it is the identification of a new state in $^{34}$Ar that has the most significant impact on the astrophysical $^{33}$Cl$(p,\gamma)$ reaction, rather than the specific assignment choice between $1^+$ and $3^-$.

A comparison of the presently determined $^{33}$Cl$(p,\gamma)$ reaction rate with previous estimates indicates that the current rate is significantly higher than expected over the temperature range of classical novae [13]. Moreover, the new identification of proton-unbound levels, together with firm spin-parity assignments, has reduced uncertainties in the rate by several orders of magnitude (a complete quantitative description will be presented later in a full paper). Consequently, in order to assess the astrophysical implications of the new reaction rate, we performed a series of nova outburst simulations using the hydrodynamic, Lagrangian, time-implicit code SHIVA [26,27]. Energy generation from nuclear reactions was obtained using a network that contains 120 nuclear species (from $^1$H to $^{48}$Ti), linked through 630 nuclear processes, with updated reaction rates from the STARLIB database [28], and we considered a representative case of an accreting 1.35 $M_\odot$ white dwarf, with characteristic values for its initial luminosity ($10^{-2}\ L_\odot$) and mass-accretion rate ($2 \times 10^{-10}\ M_\odot\ \mathrm{yr}^{-1}$). All simulations resulted in the ejection of about $5 \times 10^{-6}\ M_\odot$ of nuclear-processed material, after achieving a peak temperature of ~0.3 GK. Focusing on the ejected $^{32}$S/$^{33}$S ratio, our present results indicate a range of ~65–80. This is in contrast to the $^{32}$S/$^{33}$S ratio of ~130–200 predicted for type-II supernovae [29], and confirms the suggestion of Parikh *et al.* [9] that sulfur isotopic abundances may be used as a unique identifier of novae nucleosynthesis. Intriguingly, the recently discovered grain, Ag2_6 [30], which has been reported to have a $^{32}$S/$^{33}$S ratio of $121 \pm 30$, may now represent the most propitious candidate for originating from a classical nova explosion [31]. However, it is important to note that the best candidate nova grains appear to be sufficiently mixed with solar material that they exhibit isotopic ratios between nova and supernova model predictions. Thus, it is not presently possible to definitively distinguish the origin. We hope that this situation will change in the future.

In summary, we have performed the first detailed $\gamma$-ray spectroscopy study of the astrophysically important nucleus $^{34}$Ar, using a powerful experimental setup that combines the advanced $\gamma$-ray tracking array, GRETINA, with the focal plane system of the FMA. We have identified 5 proton-unbound excited states in $^{34}$Ar by their $\gamma$ decays, including two $\ell = 0$ resonances in the $^{33}$Cl + $p$ system at $E_r = 217$ and 397 keV, respectively, as well as a further $\ell = 1$ resonance at $E_r = 303$ keV. The newly observed 397-keV resonance has been found to dominate the $^{33}$Cl$(p,\gamma)$ stellar reaction rate over the peak temperatures of classical novae, while the 217-keV resonance governs the rate at ~0.2 GK. Further constraints on the $^{33}$Cl$(p,\gamma)$ reaction would now require a measure of the strengths of the 217- and 397-keV resonances, and we encourage additional experiments to this end. We also strongly encourage a new, comprehensive theoretical study of nova nucleosynthesis, that incorporates all the latest experimental information and has a specific focus on reproducing a wide range of astronomical observables using realistic assumptions for initial nova conditions.

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